Studies of Ultrafast Femtosecond-Laser-Generated Strain Fields with Coherent X-rays

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Abstract. In its 324 bunch-mode of operation, the Advanced Photon Source (APS) has opened new avenues of femtosecond-laser science and techniques. In this new mode, if one uses the tightly focused low-pulse energy (nJ), high repetition rate fs-laser Ti:sapphire oscillator (88 MHz) on beamline 7ID, every laser pulse and X-ray bunch can be overlapped and delayed with respect to each other, resulting in a high-repetition rate pump-probe experiment that uses all the APS X-ray bunches. This paper describes an example of how coherent X-ray experiments may be used to study laser-generated strain fields in semiconductors. With an oscillator beam focused to 6 μm onto GaAs, we have observed coherent X-ray diffraction patterns with a high-resolution camera. We have developed two techniques to observe the strain field, a topographic technique and a coherent diffraction technique. The topographic technique is quite useful to achieve a coarse spatial overlap of the the laser and X-ray beams. The coherent X-ray technique allows one to push the alignment to a few microns. This paper focuses solely on the latter technique. This experiment may help to develop techniques that will be used at the future free electron laser sources, where coherent and pump-probe experiments can be done simultaneously.

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The future is very bright for coherent X-ray and ultrafast femtosecond (fs) laser experiments at the future X-ray free electron laser (FEL) facilities. The Linac Coherent Light Source (LCLS), for example, will provide 10 orders of magnitude higher instantaneous brightness than the Advanced Photon Source (APS) and a 100 fs pulse duration. It will be transversely coherent, so pump-probe speckle experiments can be conducted routinely. Much work needs to be done to develop new techniques that will open the future science at FEL facilities. With the APS 324-bunch mode, one should be able to use the tightly focused 7ID-D fs-laser oscillator beam (1 nJ/(5μm)^2 = 4 mJ/cm^2) running at the same frequency of the ring (88 MHz) to develop a laser-pump X-ray-probe experiment, where every X-ray bunch is used. By varying the time delay between the arrival of the laser and X-ray beams, one can study time-resolved diffraction and spectroscopy on a time scale as short as the 100 ps bunch length duration of the APS. Using all the X-rays bunches of the APS allows one to consider experiments where the X-ray cross section is weak or the sample is disordered. Thus provided that significant laser excitations can be produced with a focused nJ per pulse, one can use all the known existing X-ray techniques to study these excitations. In recent work performed on the APS 7ID beamline, an amplified kHz-fs-laser pulse was shown to generate strain fields that modulate Bragg and Laue diffraction [1, 2]. In these studies, laser-pulse energy densities around ten mJ/cm^2 generate coherent acoustical phonons that propagate mostly in one dimension. The work is done in a geometry where the laser beam diameter is much larger than the laser penetration depth, and the excitation is launched impulsively due to the short pulse length. Experiments with a Ti:sapphire laser oscillator will likely be in the weak excitation limit for semiconductors with its peak 4 mJ/cm^2. But using a tightly focused laser beam would allow one to probe a new regime of excitation, where the focused beam diameter is on the same order of magnitude as the laser penetration depth. As shown recently with laser only work, the ratio of the laser beam size over the laser penetration depth influences the strain generation process [3]. In this paper, we show how one can observe laser-generated strain fields in GaAs with coherent X-ray diffraction (CXD).

The 7ID beamline cryogenically cooled Si (111) monochromator was set to 10 keV and detuned by 50% to reduce the third harmonic contamination. The full-width at half maximum (FWHM) of the beam energy bandpass is $\Delta E/E = 0.014\%$. To observe coherent diffraction patterns, the X-ray path-length difference in the sample, Δx, must be smaller than the longitudinal coherence length of the incident X-rays $l_l = \lambda/(\Delta E/E)$, where λ is the X-ray wavelength. In the Bragg diffraction case, one can show that the longitudinal coherence is preserved when $\Delta x = 2l\sin^2\theta_B < l_l$ [4]. Here θ_B is the Bragg angle, and l is the X-ray penetration length, limited by a combination of X-ray absorption and X-ray extinction effects. For perfect crystals of GaAs, we use the X-ray extinction length. For GaAs (111) and (002) at

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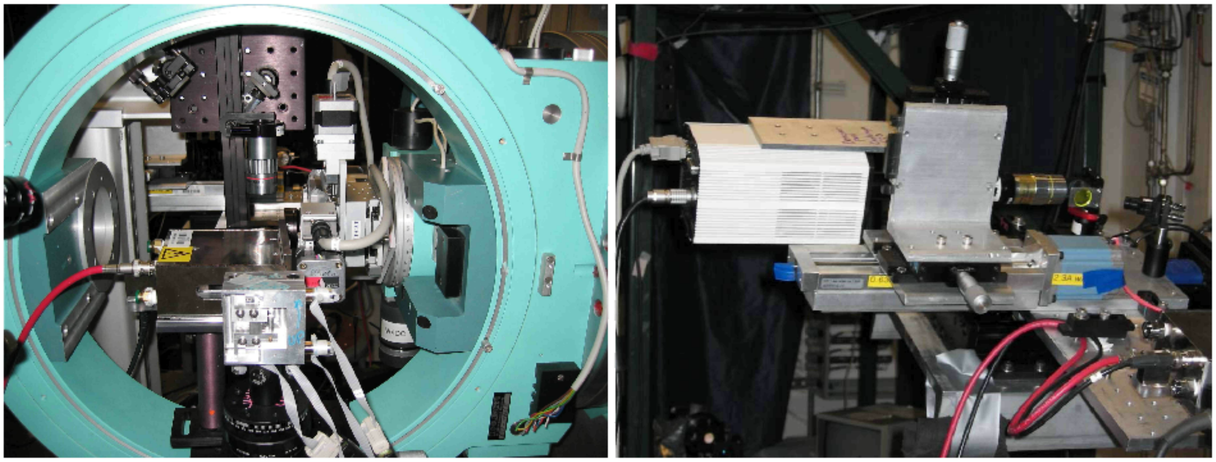

FIGURE 1. A) (left) Upstream view of the experimental set up. B) (right) Detector arm set up.

10 keV, the Bragg angles are 10.948 and 12.668 degree respectively, and l is 1.8 and 24.2 μm; thus Δx is 130 nm and 2.3 μm, respectively. Since at 10 keV, $l_l = 0.89$ μm, the longitudinal coherence condition is satisfied for GaAs (111), but not for the (002) reflection, the contrast in coherent diffraction imaging experiment for the (002) reflection will be reduced from unity.

To observe speckle in a coherent diffraction experiment, the illuminated area must also be smaller than the transverse coherence area of the source. An X-ray slit (seen in Fig. 1a) placed 20 cm from the sample can reduce the illumination area to a beam of 3×10^7 coherent X-rays/s within an area of 7 μm by 7 μm. The Ti:sapphire oscillator beam has a nominal wavelength of 800 nm, a pulse length of 30 fs, a repetition rate of 88 MHz, and an average power of 0.45 W. During the span of a week, the output power dropped to about 0.3 W. It was focused down on a GaAs (111) or (001) sample near normal incidence using a long-working-distance Mitutoyo objective with X5 magnification (see Fig. 1a). The transmission of the mirrors in the optical path and the microscope lenses reduced the delivered power to the sample by a factor 0.3, thus the maximum deliverable power on the sample was 135 mW. A set of neutral density filters was used to reduce the power on the sample. The laser focal spot could be displaced on the sample with a stepper-motor-controlled XYZ positioner (three Newport MFN stages). The sample could also be displaced in the center of a 4-circle Huber diffractometer using an Kohzu XYZ stage, controlled by stepper motors.

The diffracted X-rays were absorbed on a YAG:Ce single-crystal screen, generating visible fluorescence at 550 nm, which was imaged using a CoolSNAP HQ CCD camera and a X10 microscope objective (see Fig. 1b). The 12 bit camera is linear and the X-ray resolution of the set up is around 3 μm [5]. The response of the YAG was calibrated against the calculated flux from an ion chamber current following the approach described in Ref. [5]. One analog to digital unit of the CCD corresponds to 51.5 X-rays. The sample to YAG distance was approximately 0.65 m.

The laser focal waist was measured with scans of a razor-blade position mounted on the Kohzu stage, transverse to the laser propagation direction. The signal from a laser power meter recorded the transmitted flux. Figure 2a shows the transmitted laser signal as a function of the blade position at the optimal focal distance. The data are well fit to an error function. The Gaussian beam profile has a FWHM of 6 μm. The position of the best focus was found by displacing the blade along the laser propagation direction. Figure 2b shows a plot of the depth of focus of the microfocused laser beam. The data are well fit to a quadratic function $y = ax^2 + w_0$, where w_0 is the best focal waist, and $a = 510$ μm/mm^2. Defining the depth of focus Δf when $y = 2w_0$, $\Delta f = \sqrt{w_0/a} = 0.108$ mm. The manufacturer specifies a depth of field of 14 μm, which is a factor 8 smaller than our measurement.

Figure 3a shows a coherent diffraction pattern of Si (111) at the peak of its rocking curve. The vertical direction is parallel to the angle 2θ, while the horizontal axis records an horizontal angle $2\theta_H$. The coherence-defining slit was set to (7 μm)^2. From the fringe visibility, it is obvious that beamline 7ID has enough transverse and longitudinal coherence to perform coherent diffraction imaging on low-order Bragg reflections of semiconductors. The imaging detector provides ample resolution to resolve the fringes.

Figure 3b shows three snapshots of the bunch current in 324 bunch mode recorded by the APS accelerator division. Most of the buckets are emptied. Note the large current fluctuations for the filled buckets. We typically measure that the pulse-to-pulse stability of the oscillator power is about one percent. Since we excite our sample at the same pulse

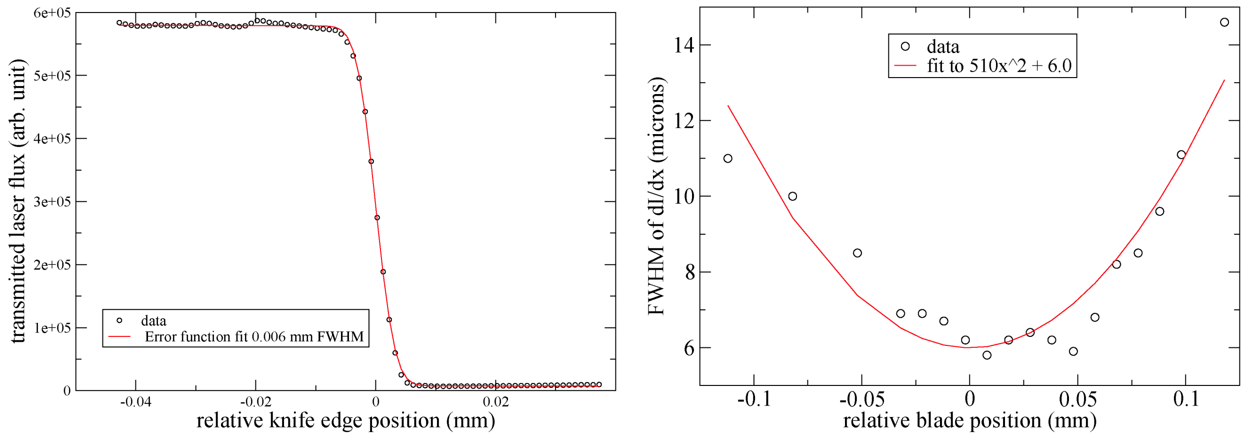

FIGURE 2. A) (left) Focal waist scan versus knife-edge transverse position. B) (right) FWHM focal waist versus knife-edge longitudinal position.

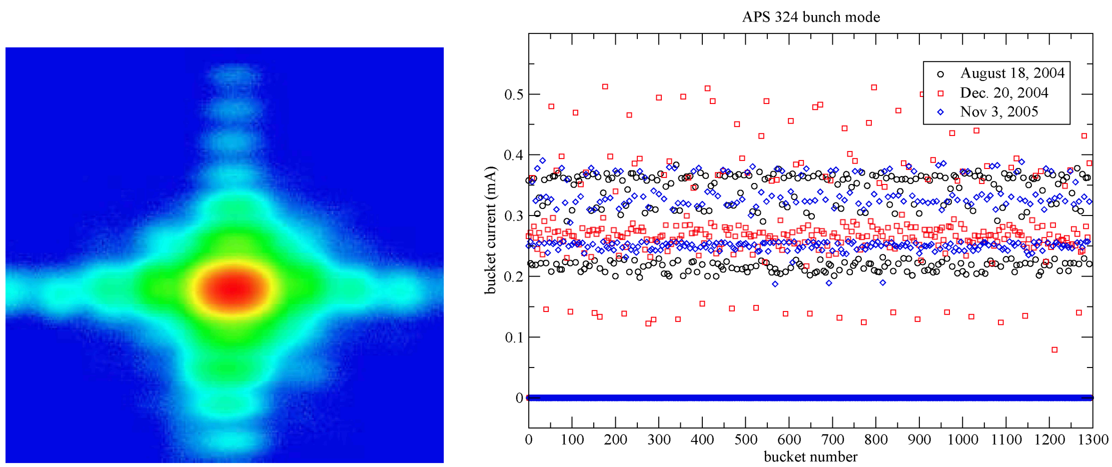

FIGURE 3. A) (left) Fraunhofer diffraction pattern of a coherent beam diffracted by Si (111) with the laser off. B) (right) The bunch current per filled bucket in 324 bunch mode for three fill snapshots.

energy per pulse, we do not expect these current variations to affect our measurements. The spread in bunch current implies a spread in bunch duration through the cube root of the bunch current.

Figure 4 shows the GaAs (002) reflection with laser on and laser off. It is interesting to note that the laser-off pattern does not show any diffraction fringes. This is due to the long optical path difference for this nearly forbidden reflection. Turning the laser on reduces the peak intensity by a factor five. The ratio of integrated intensity between images taken with the laser on and off at the peak of the rocking curve is 38%; thus the laser reduces the integrated intensity by more than a factor 2. The laser-on data were obtained with an incident power on the sample of 68 mW; 71.4% of the power is absorbed in the sample into a 6 μm FWHM Gaussian spot. The incident and absorbed peak energy density per pulse are, respectively, 1.9 and 1.35 mJ/cm^2.

In Ref. [1] and [2], fluences were reported in the range of 5 to 12 mJ/cm^2 with an amplified kHz system. Our data were obtained with a neutral density filter with an optical density of 0.3. Without the neutral density filter, the power was 2.1 times higher, and we noticed a permanent burned-in low-intensity spot on the sample's X-ray diffraction in the location of the laser spot. We did not investigate the structure of the damaged region.

Clearly, the tightly focused oscillator beam generates a strong strain field in GaAs that splits the diffracted beam in three peaks in Fig. 4b and 4c. The angle $\Delta 2\theta_B$ between the two large peaks in Fig. 4c is 75 μrad. The angle corresponds to a strain of $\Delta a/a = \Delta\theta/\tan\theta = 1.7 \times 10^{-4}$. From the thermal expansion coefficient of GaAs (5.73×10^{-6}/K), one deduces a temperature difference of 30 K. A finite element analysis calculation predicts a peak temperature rise of

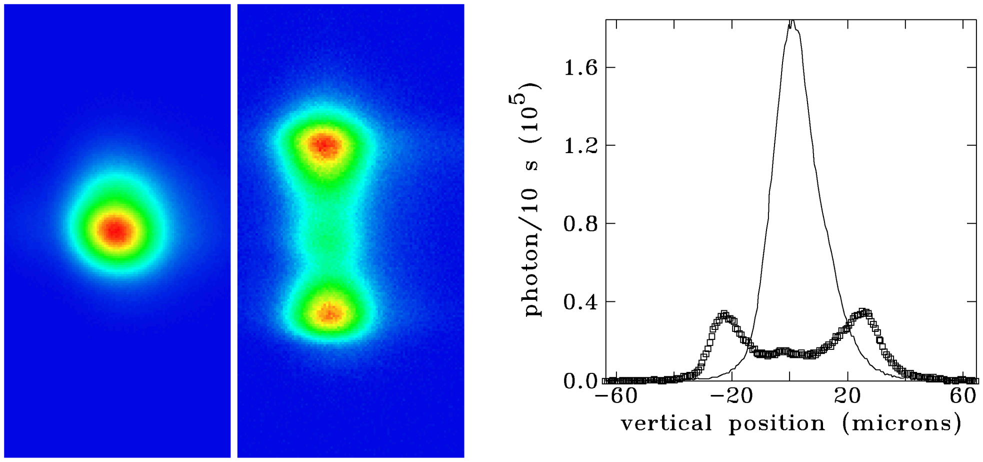

FIGURE 4. A) (left) GaAs (002) coherent diffraction pattern with laser off. The data are displayed with a rainbow color map from 0 to 1.8×10^5 ph/10s. The effective pixel size is 0.65 μm. B) (middle) GaAs (002) diffraction pattern with laser on, displayed with a color map from 0 to 3.7×10^4 ph/10s. C) (right) Vertical slices of the previous two images. The laser-off data are displayed with a solid line, while the laser-on data are shown with squares.

137 K. Since the X-ray beam footprint on the sample is 36 μm, the X-ray probe is much wider than the laser-beam spot size, thus a temperature rise of 30 K is reasonable. We did not find any notable response of the sample to the laser and X-ray time delay or to the laser pulse length.

A quantitative analysis of the microdiffraction patterns is underway to understand the microstructure of the sample. The results from the weak reflection GaAs (002) show that it should be easy to record coherent X-ray diffraction patterns following fs-laser excitation on strong reflections of semiconductors like Ge, and InSb (111) [1]. Future experiments will try to improve the spatial overlap of the X-rays with the laser focus by using microfocused X-rays. Studies of thin films will help to better match the X-ray and laser penetration depth.

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